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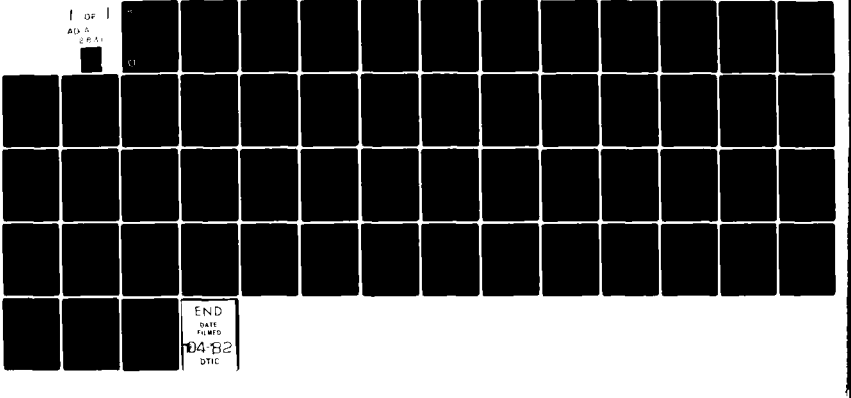

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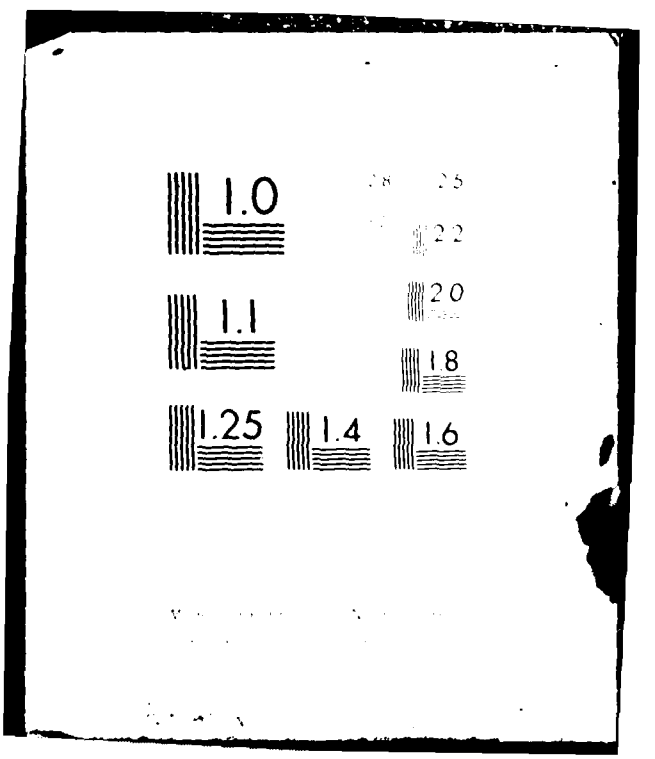
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PROPERTIES OF DUST AS AN ELECTRON AND ION ATTACHMENT SITE FOR USE IN D REGION ION CHEMISTRY

OCTOBER 1981

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Numerous observations of D region electron and ion density profiles have been difficult to interpret with chemical models based upon our present knowledge of a simple gas-phase chemistry. Often the presence of submicrometer sized particulates has been tentatively suggested as a cause of these enigmatic properties of the D region. The numerous observations of noctilucent clouds and mesospheric scattering layers are evidence for particulate matter in the D region, although the origin, composition, and size distribution of such particulates can only be estimated within rather broad ranges. This report

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20. ABSTRACT (cont)

establishes some basic properties of single particulates such as the electron-particulate and ion-particulate attachment rates, recombination rates, and photodetachment rates. A derivation of attachment coefficients for particulates acting en masse in the D region plasma is next examined. The limitations to this approach are outlined along with a discussion of preferred charge loading distributions on the particulates under various ambient conditions. Finally, illustrative examples are given of the effect of particulates on D-region electron density profiles and charge balance.

CONTENTS

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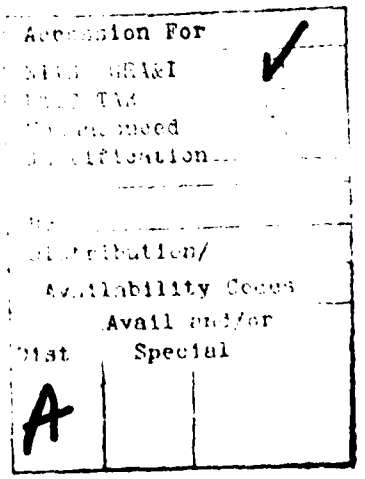

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NOTATIONS

a	Radius of dust particles, centimeters
z	Number of electronic charges on dust, a positive or negative integer
e	Electronic charge
T	Absolute temperature
$\bar{C}$	Mean thermal speed of ions or electrons. For ions, assume a mass of 50 amu. Therefore $\bar{C}_+ \approx 2.6 \times 10^4$ cm s^{-1}. For electrons, $\bar{C}_e \approx 300\ \bar{C}_+$.
F	Geometric modification shape factor
N_e	Number density of electrons, $centimeter^{-3}$
N_+	Number density of positive ions, $centimeter^{-3}$
N_-	Number density of negative ions, $centimeter^{-3}$
N_D	Number density of dust, $centimeter^{-3}$
ν_z	Frequency of capture of positive ions by dust particles with a charge z, $second^{-1}$
ν_z^e	Frequency of capture of electrons by dust particles with a charge z, $second^{-1}$
ν_z^-	Frequency of capture of negative ions by dust particles with a charge z, $second^{-1}$
ν_{ph}	Photodetachment frequency, $second^{-1}$
P_z	Fraction of dust particles which carry a charge z
k_D^e	Coefficient of attachment of an electron to a single dust particle, $centimeter^3\ second^{-1}$
k_D^+	Coefficient of attachment of a positive ion to a single dust particle, $centimeter^3\ second^{-1}$
K_D^e	Effective attachment coefficient of electrons in a distribution of dust particles, $centimeter^3\ second^{-1}$
K_D^+	Effective attachment coefficient of positive ions in a distribution of particles, $centimeter^3\ second^{-1}$

1. INTRODUCTION

The D region is the lowest portion of the ionosphere and nominally encompasses the altitude range of 60 to 90 km. It represents a transition region for the ionospheric plasma composition. Above 80 to 85 km, daytime composition is limited to molecular ions such as NO^+ and O_2^+; while below 80 to 85 km, the simple molecular ions enter into clustering reactions which convert the positive ion composition into one dominated by water clusters. Electrons also undergo attachment processes in the D region to form negative ions; by day this attachment appears to take place below 70 to 75 km. By night it occurs as high as 90 km, but there are still many unanswered questions concerning the negative ion chemistry.

The sources of ionization within the D region are the "tail end" of the solar x-ray spectrum, mainly at wavelengths less than 1 nm and in the 3- to 10-nm range, and two strong solar emission lines, Lyman beta at 102.6 nm which ionizes O_2 and Lyman alpha at 121.6 nm which ionizes NO. Ionization from these sources occurs principally above 75 km. Below 70 km galactic cosmic rays are the main (though small) source of ionization. At higher latitudes charged particle precipitation and associated bremsstrahlung x-rays may occasionally be the dominant ionization source.

The deposition of micrometeors, at the altitude of the lower portion of the E region and upper portion of the D region, produces a concentration of metallic ion layers at 90 to 110 km and occasional occurrences of sporadic E reflection. Estimates have shown that this micrometeor flux may be sufficiently large to serve as source material for the mesospheric scattering layers and noctilucent clouds which are often observed in the 80 to 85 km range.[1]

Certain D region phenomena have been observed which have no ready explanation in terms of conventional ion chemistry; for example, electron density profiles have occasionally shown layers of sharply decreased concentration in the 80 to 90 km range, which could be interpreted as due to particulate layers which have the ability to attach electrons.[2] This ability would suggest, therefore, that there is a large group of primarily negatively charged species, either

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[1]D. M. Hunten, R. P. Turco, and O. B. Toon, 1980, "Smoke and Dust Particles of Meteoric Origin in the Mesosphere and Stratosphere," J Atmos Sci, 37:1342.

[2]A. Pedersen, J. Troin, and J. A. Kane, 1970, "Rocket Measurements showing a Removal of Electrons Above the Mesopause in Summer at High Latitude," Planet Space Sci, 18:945-947.

ions or particulates, present in the upper D region.[3,4] And indeed, conductivity measurements throughout the stratosphere and mesosphere give evidence of light and "massive" ions, which may be interpreted to be charged particulates. D-region electron densities observed during some solar eclipses have indicated a surprisingly rapid electron density decrease which cannot be explained on the basis of current knowledge of electron-ion rates of recombination.[5,3]

With these examples of anomalous behavior in mind, the purpose of this report is to establish properties of particulates necessary for use in D-region predictive computer codes. These properties are electron-particulate and ion-particulate collision frequencies, attachment and recombination rates, and electron photodetachment rates. To achieve this result, the coefficients will be examined on three levels: (1) Coefficients will be derived for single particles impacting with electrons or ions. (2) An "effective" or bulk attachment coefficient shall be derived which pertains to a population of dust mixed with ambient electrons and positive ions. This derivation follows the work of Parthasarathy and Rai[6] and Parthasarathy[7] but has certain limitations which shall be examined. This treatment is instructive since it derives the charge loading on a population of dust and outlines the sensitivity of the attachment coefficients to various parameters. (3) Particulates will be included and evaluated as electron or ion attachment sites within a full time-dependent chemical code which includes all other competitive processes of the D-region chemistry.

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[3]M. G. Heaps, F. E. Niles, and R. D. Sears, 1978, Modeling the Ion Chemistry of the D-region: A Case Study Based Upon the 1966 Total Solar Eclipse, ASL-TR-0015, US Army Atmospheric Sciences Laboratory, White Sands Missile Range, NM.

[4]M. G. Heaps and J. M. Heimerl, 1980, "The Quiet Midlatitude D-region: A Comparison Between Modeling Efforts and Experimental Efforts," J Atmos Terr Phys, 42:733.

[5]E. A. Mechtly, C. F. Sechrist, Jr., and L. G. Smith, 1972, "Electron Loss Coefficients for the D-region and the Ionosphere from Rocket Measurements During the Eclipses of March 1970 and November 1966," J Atmos Terrs Phys, 34:641

[6]R. Parthasarathy and D. B. Rai, 1966, "Effect of Meteoric Dust on the Effective Recombination Coefficient in the Lower Ionosphere," Radio Sci, 1:1401.

[7]R. Parthasarathy, 1976, "Mesopause Dust as a Sink for Ionization," J Geophys Res, 81:2392.

2. PROPERTIES OF MESOSPHERIC PARTICLES

Noctilucent clouds (NLC) have long provided the most direct and visual evidence that particulates, most likely pure ice or ice coated dust, tend to layer at the summer mesopause. The rare observation of NLC during stratospheric warmings indicates layering in winter also.[8] OGO-6 satellite observations[9] indicate that particulate layers near the 82 km upper bound of the mesosphere may be a persistent high latitude feature, while global lidar observations show a sodium layer in this same region. Witt[10] and Fogle and Rees[11] have calculated NLC particulate sizes to be of the order of 0.1μm to 0.2μm in radius with number densities of the order of 1 cm^{-3}. Donahue et al[9] also estimate that 0.13μm radius particles with number densities of 15 to 40 cm^{-3} would successfully explain their scattering observations. An earlier work by Divari[12] based upon rocket samples states that particulate concentrations in the mesosphere are an inverse function of size down to 0.05μm. Also model calculations by Hunten et al[1] indicate a similar inverse size and concentration relationship in the 80- to 90-km altitude range for particles less than 0.001μm.

There are three alternatives for the origin of NLC particulates: (1) They are terrestrial and ascend by the action of turbulence, (2) they are formed from in-situ ion clusters, or (3) they are meteoritic.

The first possibility seems unlikely considering the insufficient magnitude of the eddy diffusion coefficient in the stratosphere and mesosphere (see, for example, Hunten[13]). As for the second possibility, Witt[14] proposed that water

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[8]R. A. Hamilton, 1964, "Observations of Noctilucent Clouds Near Midwinter," Met Mag Lond, 93:201.

[9]T. M. Donahue, B. Guenther, and J. E. Blamont, 1972, "Noctilucent Clouds in the Daytime: Circumpolar Particulate Layers Near the Summer Mesopause," J Atmos Sci, 29(6):1205.

[10]G. Witt, 1960, "Polarization of Light From Noctilucent Clouds," J Geophys Res, 65:925-933.

[11]B. Fogle and M. Rees, 1972, "Spectral Measurements of Noctilucent Clouds," J Geophys Res, 77:720.

[12]G. Divari, 1964, "The Dust Concentration in the Upper Layers of the Earth's Atmosphere," Geomagnetism and Aeronomy, 4:688-692.

[1]D. M. Hunten, R. P. Turco, and O. B. Toon, 1980, "Smoke and Dust Particles of Meteoric Origin in the Mesosphere and Stratosphere," J Atmos Sci, 37:1342.

[13]D. M. Hunten, 1975, Vertical Transport in Atmospheres, Atmospheres of Earth and the Planets, D. Reidel Publishing Co., Dordrecht-Holland, pp 57-72.

[14]G. Witt, 1969, "The Nature of Noctilucent Clouds," Space Res, IX:157-169, North Holland Publishing Co.

cluster ions in the mesosphere[15] [16] serve as condensation or sublimation nuclei. Such a clustering mechanism has been investigated for aerosols in general.[17] [18] However, the mechanism whereby clusters of ten or fewer molecules could grow or combine to form the size of particulates nominally present in the D region (hundreds or thousands of molecules) has not yet been identified. Assisted by a model calculation, Reid[19] has argued that if a substratum of small particulates of nominal 0.001μm size exists, then the growth of ice needles of a detectable size (0.13μm) is possible under constraints of mesospheric temperatures and low water vapor. The idea that meteoritic dust particles could be sufficiently numerous to form layers dense enough for NLC also has been theoretically supported by Chapman and Kendall.[20] Hunten et al[1] showed that a nominal meteor flux of 10^{-15} g cm^{-2} s^{-1} is sufficient to provide a source of particulates with sizes of the order of 0.001μm and number densities of 10^3 cm^{-3}. Also the source of particles may be multiple and seasonally dependent as suggested by data of Clemesha et al.[21] Thus, mesospheric particulate sizes and their number densities can apparently range from an upper bound of 0.1μm radius with number densities on the order of 10 cm^{-3} or less, to a lower bound of 0.001μm sized particulates with number densities on the order of 10^3 cm^{-3}. This range of concentrations and sizes will be used as representative in the subsequent calculations and modeling in this report.

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[15]R. D. Narcisi and A. D. Bailey, 1965, "Mass Spectrometric Measurements of Positive Ions at Altitudes From 64 to 112 Kilometers," J Geophys Res, 70:2687.

[16]F. Arnold and W. Joos, 1979, "Rapid Growth of Atmospheric Cluster Ions at the Cold Mesopause," Geophys Res Letters, 6:763-766.

[17]A. W. Castleman, Jr., 1974, "Nucleation Processes and Aerosol Chemistry," Space Sci Rev, 15:547-589.

[18]A. W. Castleman, Jr., 1979, "Nucleation and Molecular Clustering About Ions," Adv in Colloid and Interface Sci, 10:73-128.

[19]G. C. Reid, 1975, "Ice Clouds at the Summer Polar Mesopause," J Atmos Sci, 32:523.

[20]S. Chapman and P. C. Kendall, 1965, "Noctilucent Clouds and Thermospheric Dust: Their Diffusion and Height Distribution," Quart J Roy Met Soc, 91:115.

[1]D. M. Hunten, R. P. Turco, and O. B. Toon, 1980, "Smoke and Dust Particles of Meteoric Origin in the Mesosphere and Stratosphere," J Atmos Sci, 37:1342.

[21]B. R. Clemesha, V. W. J. H. Kirchoff, D. M. Simonich, and H. Takahashi, 1978, "Evidence of an Extra-Terrestrial Source for the Mesospheric Sodium Layer," Geophys Res Letters, 5:873-877.

3. ELECTRON AND ION ATTACHMENT TO SINGLE DUST PARTICLES

The first quantitative steps towards examining the properties of dust in the presence of charged particles were taken in 1960 by Natanson,[22] who formulated a kinetic approach to the charging of dust by electrons and ions. Parthasarathy and Rai[6] used Natanson's approach to derive attachment coefficients for dust particles with electrons and positive ions in the mesosphere. This work was extended by Parthasarathy[7] to include a probabilistic formulation of the basic equilibrium equations including an explicit photodetachment term.

A dust particle is charged when an electron or ion collides with and is captured by the particle. The capture frequency, ν, equal to the collision frequency (with a collision efficiency of 1), may be expressed as

$$\nu = \pi a^2 N \bar{C} F \quad (s^{-1}) , \tag{1}$$

where πa^2 is the geometric cross section of the particle, N is electron or ion density, $\bar{C}$ is mean velocity, and F is a capture rate modification function or "shape factor" which modifies the geometric cross-section and whose value depends upon the resident charge of the particle and its radius. This shape factor, F, expresses the attractive (or repulsive) effects of charged dust particles to approaching electrons or ions (figure 1).

Several points should be noted. First, the F factor for uncharged dust particles enhances the geometric cross section, an effect which increases as the particle shrinks. This effect is due to the setting up of an image charge within the particle as the electron or ion approaches its surface. Second, powerful repulsive effects lead to a practical limit of one charge for particles of approximately 0.01μm in size or less. For such small particles, recombination or neutralization becomes greatly enhanced as the particle becomes charged. Only for particles larger than 0.1μm does multiple charging become probable.

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[22]G. L. Natanson, 1960, "On the Theory of the Charging of Amicroscopic Aerosol Particles as a Result of the Capture of Gas Ions," Sov Phys Tech Phys, (English Translation), 5:538.

[6]R. Parthasarathy and D. B. Rai, 1966, "Effect of Meteoric Dust on the Effective Recombination Coefficient in the Lower Ionosphere," Radio Sci, 1:1401.

[7]R. Parthasarathy, 1976, "Mesopause Dust as a Sink for Ionization," J Geophys Res, 81:2392

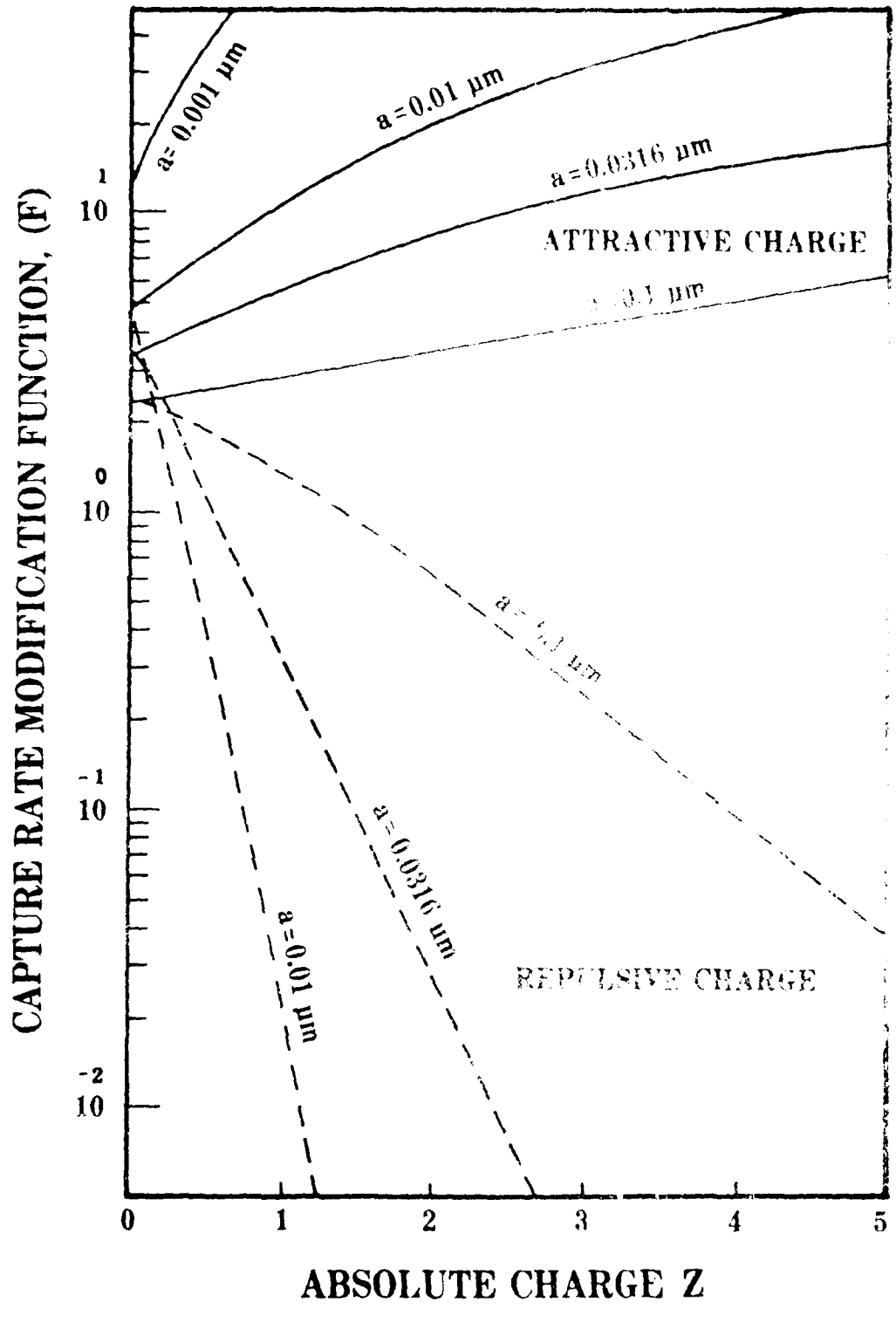

Figure 1. A plot of the capture rate modification function (F) against the absolute charge, Z, on a dust particle of radius 0.1μm to 0.001μm. The value of the capture rate modification function expresses the ratio of the "effective" capture cross section of a dust particle to the simple geometric cross section and is dependent upon electrostatic effects.

The dust charge attachment coefficient, k_D, for an isolated particle is defined as

$$k_D = \nu/N = \pi a^2 \bar{C} F \quad (cm^3 s^{-1}) , \qquad (2)$$

where k_D is a function of particle size, resident charge, electron or ion mass, and velocity. k_D is to be treated as a standard chemical rate constant, where the term in the set of continuity equations becomes $k_D^e [N_D] [N_e]$ or $k_D^+ [N_D] [N_+]$. Table 1 lists representative values of the coefficients k_D^e and k_D^+ for a nominal mesospheric temperature T = 150°K.

Points which are apparent from table 1 are: (1) the smaller the dust, the more rapidly the electron attachment coefficient decreases with increasing negative charge, (2) the larger the particle, the greater tendency it will have to attach electrons and, hence, provide an electron sink. A single large particle (that is, radius >0.1 μm) is therefore as effective as several smaller particles in perturbing the charge balance.

TABLE 1. k_D^e AND k_D^+, ELECTRON AND POSITIVE ION ATTACHMENT COEFFICIENTS FOR A SINGLE DUST PARTICLE AT T = 150°K

Dust Radius (μm)	Charge (e)	k_D^e ($cm^3 s^{-1}$)	k_D^+ ($cm^3 s^{-1}$)
	Z		
0.001	+1	2.6(-5)	1.3(-33)
	0	3.3(-6)	1.0(-8)
	-1	4.2(-31)	8.4(-8)
	-2	2.3(-37)	1.6(-7)
0.01	+1	2.8(-4)	7.3(-11)
	0	1.2(-4)	3.9(-7)
	-1	2.3(-7)	9.1(-7)
	-2	1.1(-10)	1.7(-6)
0.1	+1	5.0(-3)	1.1(-5)
	0	5.5(-3)	1.7(-5)
	-1	3.4(-3)	1.5(-5)
	-2	1.3(-3)	2.4(-5)
	-3	5.2(-4)	3.2(-5)
	-4	2.1(-4)	4.0(-5)
	-5	8.3(-5)	4.9(-5)
	-6	3.3(-5)	5.7(-5)
	-7	1.3(-5)	6.5(-6)
	-8	5.2(-6)	7.4(-6)

A further insight may be provided by a plot (figure 2) comparing the magnitude of the attachment coefficients, k_D^e, for z = -1, 0, and +1 charged dust, as a function of particle radius, and the electron-ion recombination coefficients for NO^+ and water cluster ions. This figure shows that although normally the most important loss mechanism for electrons is recombination to cluster ions, electron attachment to particulates could be dominant if the dust number density were of the same magnitude as the ion number density and (a) dust were positively charged, (b) dust were neutral but larger than 0.002 μm, or (c) the dust particle had a single negative charge and exceeded 0.01 μm in radius. The inferences of dust size distribution in the mesosphere allow that there could be much dust at sizes of 0.01μm or smaller. In that case the probability of mesospheric dust affecting the D-region charge balance would be considerable through the formation of an electron sink.

4. EFFECTIVE CHARGE ATTACHMENT COEFFICIENTS

4.1 Derivation

The previous section explicitly derived the charge attachment frequency for an isolated dust particle of a given size and a known resident charge. However, in the D region, a population of dust particles will tend to possess a distribution of charge which will be established by the competitive attachment and detachment of electrons and ions. Parthasarathy and Rai[6] and Parthasarathy[7] initially addressed the calculation of the attachment coefficients for dust "en masse." In this section we analyze the limitations of this approach but show that it provides knowledge of the resident charge distribution function of dust for various ambient conditions. Photodetachment is also included in this formulation as an explicit term, and suitable experimental values are examined.

Parthasarathy[7] stipulates that the equilibrium condition of dust mixed with charged species is considered to be a statistically stationary distribution. This condition is formulated by stating that the frequency at which the fraction of dust particles with a charge z is being depleted by ion capture, electron capture, and photodetachment is equal to that frequency at which the fraction of dust particles with z + 1 charges are acquiring electrons or negative ions plus the frequency at which the fraction of dust particles with z - 1 charges are acquiring positive charges and suffering photodetachment of electrons (after Parthasarathy[7]).

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[6]R. Parthasarathy and D. B. Rai, 1966, "Effect of Meteoric Dust on the Effective Recombination Coefficient in the Lower Ionosphere," Radio Sci, 1:1401.

[7]R. Parthasarathy, 1976, "Mesopause Dust as a Sink for Ionization," J Geophys Res, 81:2392.

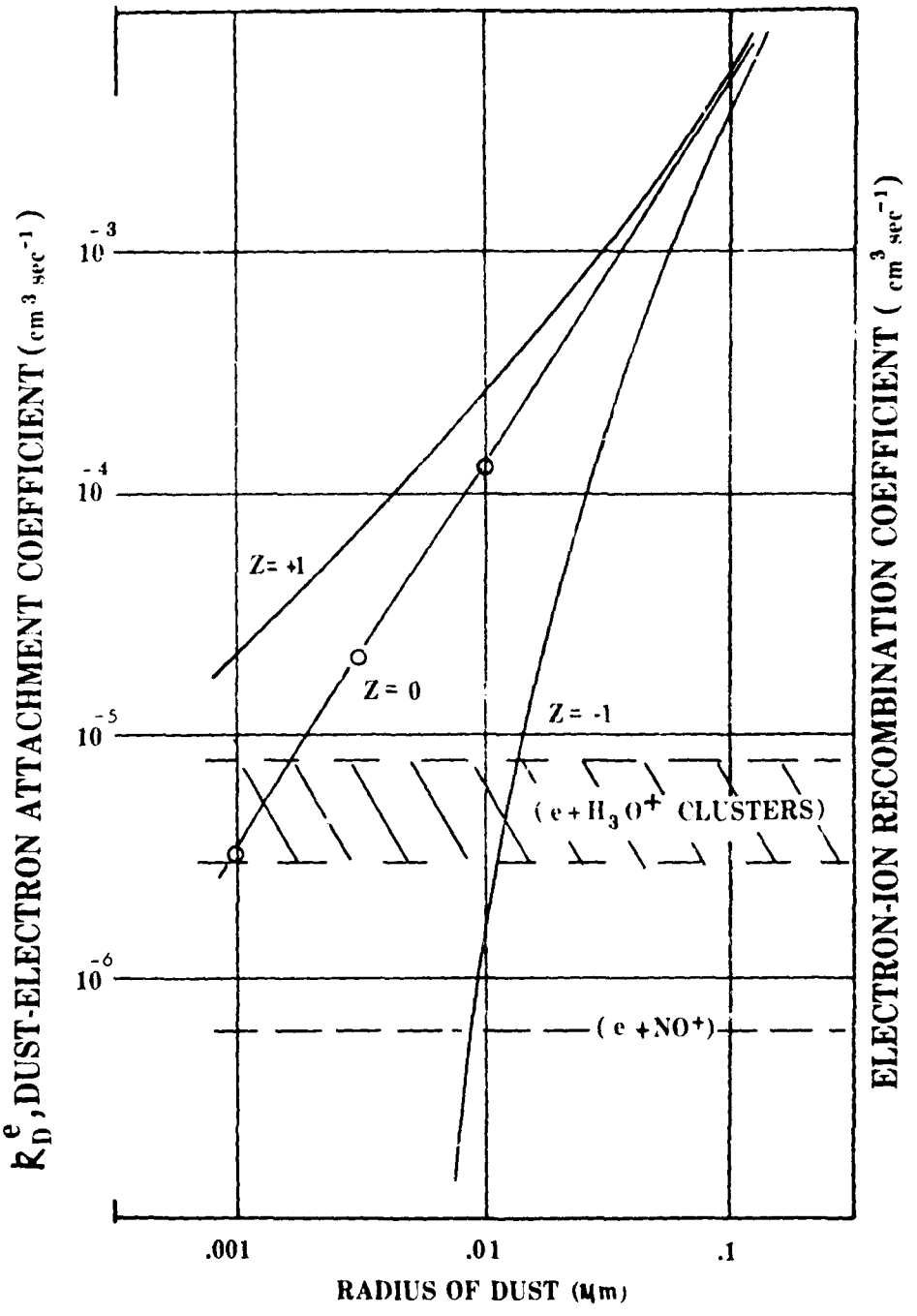

Figure 2. A plot showing the magnitude of k_D^e, the dust-electron attachment coefficient (left ordinate), for various radii of dust of charge Z = -1, 0, +1. For comparative purposes the electron-ion recombination coefficient for nitric oxide ions and water cluster ions is plotted on the right ordinate.

Expressed algebraically,

$$P_z \left(\nu_z^+ + \nu_z^e + \nu_z^- + \nu_{ph}\right) = P_{z+1}\left(\nu_{z+1}^e + \nu_{z+1}^-\right) + P_{z-1}\left(\nu_{z-1}^+ + \nu_{ph}\right) . \quad (3)$$

By setting $P_z = 0$ for a large value of charge (say $z = -9$, meaning that no dust has 9 or more electrons on it), and solving the other P_z's in terms of P_{-8} and so forth, a recursion relation may be obtained. Then, after normalizing with $\Sigma P_z = 1$, solve for each P_z. Neglecting negative ions for the daytime D region, the effective attachment coefficients are then defined as:

$$K_D^+ = \frac{1}{N_+} \Sigma P_z \nu_z^+ = \frac{1}{N_e} \Sigma P_z (\nu_z^e - \nu_{ph}) ; \quad (4)$$

$$K_D^+ = \frac{1}{N_e} \Sigma P_z \nu_z^e - \frac{\Sigma P_z \cdot \nu_{ph}}{N_e} . \quad (5)$$

The attachment coefficient for electrons, K_D^e, is:

$$K_D^e = \frac{\Sigma P_z \nu_z}{N_e} . \quad (6)$$

Substituting equation (6) into (5) and remembering $\Sigma P_z = 1$,

$$K_D^+ = K_D^e - \frac{\nu_{ph}}{N_e} , \quad (7)$$

that is, the effective attachment coefficient for positive ions is equal to the effective attachment coefficient for electrons diminished by the photodetachment frequency divided by the ambient electron density.

This formulation incorporates the following assumptions and restrictions which limit its applicability to D region time dependent chemical codes:

a. This formulation is explicitly stationary. An equilibrium is assumed between the rate of attachment and detachment of ions and electrons for the

derivation of the P_z fractions and the coefficients. As such, these coefficients are unsuitable for use in time dependent programs designed for rapidly varying atmospheric conditions such as eclipse times, sunrise and sunset, or nuclear burst phenomena.

b. It is assumed that $N_e = \Sigma N_+$. This assumption of charge neutrality among electrons and ions is equivalent to the assumption that the number of particulates is too small to perturb the charge balance, which may be an unrealistic assumption for the mesosphere. Therefore, since the derived P_z values are functions of the N_e or ΣN_+ used, the calculation of new P_z's and the effective coefficients would have to be handled iteratively in a computer code each time new N_e and ΣN_+ were computed.

c. This formulation (equation 3) neglects competitive chemical attachment processes such as the recombination reactions between positive ions and electrons which would change P_z values and K_D^e and K_D^+.

Within these restrictions the values of K_D^e and K_D^+ derived by Parthasarathy[7] are useful for steady state chemical codes with a comparatively small number of particulates. But if no restriction is to be placed upon the size of the dust population or upon the concentrations of electrons or positive ions, then a set of stepwise reactions utilizing the coefficients for individual dust in various charge states is more appropriate (section 2). However, a benefit of the approach by Parthasarathy is to predict the range and stability of resident charge loading expected on a population of dust under various ambient conditions. Thus, the number of piecewise reactions sufficient for dust of a given size may be predicted for use in the chemical reaction code.

4.2 Choice of Photodetachment Coefficient for Dust

The photodetachment rate for the dust particle, ν_{ph}, is a key and governing parameter in these computations. Photodetachment yields of many materials are difficult to measure in a laboratory, and, in turn, whether such measurements would be valid for mesospheric conditions is difficult to assess. The best that may be done is to assume a likely composition for dust and establish a plausible range for the photoelectric yield over the incident solar wavelengths which exist in the mesosphere.

Photoelectric yield of a given material is a function of many parameters such as surface roughness, angle of incidence of photons, temperature, and possible adsorption of gases on the surface. For example, photoelectric yield will

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[7]R. Parthasarathy, 1976, "Mesopause Dust as a Sink for Ionization," J Geophys Res, 81:2392.

increase with greater roughness and angle of incidence.[23] Adsorption of gases such as molecular oxygen or hydrogen onto the surface may reduce the yield and shift the cutoff wavelength further into the ultraviolet;[23] whereas very small amounts of gases may increase yield and shift the cutoff wavelength to longer wavelengths through the formation of surface "double layers" which allow the creation of surface states about 0.4 ev below the Fermi level.[24]

If meteoritic material from galactic space, possibly ice coated, is assumed to be the constituent of the dust particles, then the photodetachment values for moon dust,[25] nickel,[23] or ice[6] would be reasonable options. Presumably, D-region meteoritic depositions would be of similar material to lunar surface dust. Willis et al[25] determined the photoelectric yield of samples of moon dust from the Apollo 14 and 15 missions. These determinations for moon dust were made at 10^{-7} to 10^{-9} torr, a more absolute vacuum than in the D region by five orders of magnitude. Impurities in the samples were baked out for 1 week at 150°C. Auger electron spectroscopy and chemical analysis yielded a composition of: 48 percent of SiO_2, 18 percent of Al_2O_3, 11 percent of [illegible], 10 percent of FeO, etc., which agrees with other determinations of composition for silicate-type meteors.[26] Peak emission of 10^{-1} electrons per photon occurs at 800 angstroms, with smaller values of 10^{-6} at 2000 angstroms and 10^{-12} at 600 angstroms. For solar flux at the top of the earth's atmosphere the composite yield is 2.8×10^{9} (πa^2) photoelectrons per second for lunar surface material, where πa^2 represents the surface area in square centimeters. Attenuation of solar flux in the Schumann-Runge Bands and Lyman α in the mesosphere reduces this photodetachment frequency of neutral dust at 80 km to about 1×10^{9} (πa^2) s^{-1}. Since iron and nickel meteorites have been commonly reported,[26] the photoelectric yield for elemental nickel was obtained in Weissler.[23] When the appropriate solar flux is used for Lyman α and the Schumann-Runge Bands attenuation at 80 km is accounted for, the resultant photodetachment for nickel is 1.5×10^{8} (πa^2) s^{-1}. Values of photodetachment for ice[6] are 1.9×10^{8} (πa^2) s^{-1}. To bracket these previous determinations for different materials and to allow for the considerable uncertainty which still remains, the following values of photodetachment frequency are chosen to

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[23]G. L. Weissler, 1956, "Photoionization in Gases and Photoelectric Emission from Solids," *in* Handbuch der Physik, Vol XXI, Springer-Verlag, 304-382.

[24]B. Feuerbacher and B. Fitton, 1972, "Photoemission from Surface States on Tungsten," Phys Rev Letters, 29(12):786.

[25]R. F. Willis, M. Anderegg, B. Feuerbacher, and B. Fitton, "Photoemission and Secondary Electron Emission from Lunar Surface Material," *in* Photon and Particle Interactions with Surfaces in Space, Proceedings of the 6th Eslab Symposium, Noordwijk, the Netherlands, 26-29 September 1972. R. J. L. Grand, Ed., D. Reidel Publishing Co., Dordrecht-Holland/Boston - USA.

[6]R. Parthasarathy and D. B. Rai, 1966, "Effect of Meteoric Dust on the Effective Recombination Coefficient in the Lower Ionosphere," Radio Sci. 1:1401.

[26]J. A. Jacobs, R. D. Russell, and J. T. Wilson, 1959, Physics and Geology, McGraw Hill, NY.

be incorporated into the calculations of this report (expressed for zero optical depth): $2.8 \times 10^{10} (\pi a^2)\ s^{-1}$, $2.8 \times 10^{9} (\pi a^2)\ s^{-1}$, $2.8 \times 10^{8} (\pi a^2)\ s^{-1}$, and 0.

Since mesospheric particles may possess a negative or positive charge, the question arises as to whether an electrostatic potential on a dust particle would modify the photodetachment yield from that dust. Manka[27] explored the result of having an electrostatic charge on the surface of dust and the resultant Coulomb retarding of the photoelectron current. He was specifically interested in dust on the lunar surface and showed, for example, that the current density of photoelectrons suffered a reduction of 26 percent in emitted electrons with a single positive charge. Our calculations are not adjusted for this small effect. In the case of a negative potential, the magnitude of the photoelectron current was not modified. That is, negatively charged dust will provide the same photoelectron current as neutral dust and obey the same wavelength dependence. Photoemission will not be enhanced.

A final comment is necessary about the differences in photoemission between single molecules and a solid composed of many molecules: It was considered possible that dust could photoemit in the visible, a fact suggested by analogy in the work of Woo et al[28] who described photoelectron detachment from the O_2^- molecule by visible wavelengths. This idea was supported by Chanin et al[29] who showed that electrons attaching to molecular oxygen to form O_2^- could possibly enter a $v = 1$ vibrational state, described by a repulsive potential energy curve, which could then lead to autodetachment. However, in the case of a solid dust particle, such surface metastable states of high energy would not exist since the energy of the impacting electron would quickly dissipate as momentum and heat through neighboring atoms. The "extra" electron causing the unbalanced negative charge on a dust particle would be indistinguishable from any other electron occupying normal bound conduction states. Photoemission would always operate against the work function of the material and exhibit the same cutoff frequency for irradiation as that for an uncharged surface. Therefore, it would not be likely for dust to photoemit in the visible.

4.3 Charge Loading on Dust

The first result of performing the calculation of the effective attachment coefficients, K_D^e and K_D^+, is the determination of the fractions of the dust

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[27]R. H. Manka, "Plasma and Potential at the Lunar Surface," in Photon and Particle Interactions with Surfaces in Space, Proceedings of the 6th Eslab Symposium, Noordwijk, The Netherlands, 26-29 September 1972, D. Reidel Publishing Co., Dordrecht-Holland/Boston - USA.

[28]S. B. Woo, L. M. Branscomb, and E. C. Beaty, 1969, "Sunlight Photodetachment of Ground State O_2," J Geophys Res, 74:2933.

[29]L. M. Chanin, A. V. Phelps, and M. A. Biondi, 1962, "Measurements of Attachment of Low Energy Electrons to Oxygen Molecules," Phys Rev, 128:219.

population which carry a particular charge. Table 2 lists the percentages of the total dust population which carry given numbers of electrons for dust of radii 0.001μm, 0.01μm, and 0.1μm under various ambient electron densities.

The tabulation reveals some of the most graphic aspects of dust behavior in the D region: For example, the charge for dust of radius 0.01μm, or smaller, is one electron per dust particle. (These results are similar to those of figures 3 and 4 in Parthasarathy.[7]) This loading holds for a wide range of ambient electron number densities of 100 to 10,000 centimeter^{-3} and photodetachment values of 0 to 10^{-1} s^{-1}. Under similar ambient conditions, dust sized midway between 0.01μm to 0.1μm holds a more or less stable charge of 2 to 3 electrons which shifts to 1 electron or no electrons at high photodetachment rates. Dust of radius 0.1μm presents a less clear-cut picture. The normal loading is a broad distribution of 5, 6, or 7 electrons which is displaced to positive values of charge where photodetachment values >1 s^{-1} and electron densities below 1000 cm^{-3} are used. In each of these cases, photodetachment does not exert an influence until it exceeds about 1 s^{-1}.

This insensitivity to photodetachment rates below a certain threshold is an important result since a rigorous number for dust photodetachment rates has been hard to obtain, and this difficulty had been an area of uncertainty in chemical models. A further result is that the smaller dust sizes will exist in the D region with only a single negative charge if they are charged at all.

As an alternate way to illustrate the competing elements of this situation, figure 3 shows a plot of photodetachment frequency for moon dust and ice along with electron attachment frequency for dust charged Z = 1, 0, +1 and frequency for the recombination of electrons with NO^+ and hydrated water cluster ions. An ambient electron and ion density of 1000 cm^{-3} has been used for this illustration.

From an examination of figure 3, a number of points are apparent:

a. Positively charged dust of any size can competitively acquire electrons when compared to normal electron-ion recombination processes. Uncharged dust of radius $\gtrsim$ 0.002μm can compete effectively for electrons even when the positive ions are water clusters (as is the case for altitudes below 80 km). Negatively charged dust can only compete effectively for electrons if the particles have a radius $\gtrsim$ 0.02μm. Dust with radii on the order of 0.1μm could attach several electrons, but because of the nominally low number densities of 0.1μm radius dust ($\sim$ 10 cm^{-3}), the overall electron attachment to dust of this size is probably of secondary importance when compared to the normal electron-ion recombination processes, a point already noted by Parthasarathy.[7] However, if dust particles of much smaller sizes were present in large numbers, as is most likely the case, then dust could appreciably alter the ambient electron and ion equilibrium densities. The precise effect

[7]R. Parthasarathy, 1976, "Mesopause Dust as a Sink for Ionization," J Geophys Res, 81:2392.

TABLE 2. PERCENT OF DUST POPULATION WITH A PARTICULAR NUMBER OF ELECTRONS FOR T = 150°K*

Dust Particle Radius (μm)		Percentage 0.001			0.01			0.1		
Ambient Electron Density		100	1000	10,000	100	1000	10,000	100	1000	10,000
Photodetachment Rate (s^{-1})	Charge (e) Z									
0	-4							7	6	6
	-5							28	28	28
	-6							40	40	40
	-7							20	20	20
	-8							3	3	3
$2.8 \times 10^{8}(\pi a^2)$	0	2	2	2		1.3	0			
	-1	97	97	97		90	88	9		
	-2					8	11	30		
	-3							36	8	
	-4							17	28	9
	-5							3	39	32
	-6								20	39
	-7								4	17
	-8									3
$2.8 \times 10^{9}(\pi a^2)$	0	22	5	2			0		2	
	-1	76	95	97			90		9	
	-2						8		30	
	-3								36	8
	-4								18	28
	-5								3	39
	-6									20
	-7									4
$2.8 \times 10^{10}(\pi a^2)$	8								1	
	7								3	
	6								6	
	5								10	
	4								15	
	3						0		19	
	2	3					0		18	
	1	19					7		14	
	0	56	22	5			90		6	1
	-1	20	76	95			2		3	9
	-2								1	30
	-3									36
	-4									17
	-5									3

*Columns may not always total 100 due to rounding and to neglecting states with less than 0.5 percent.

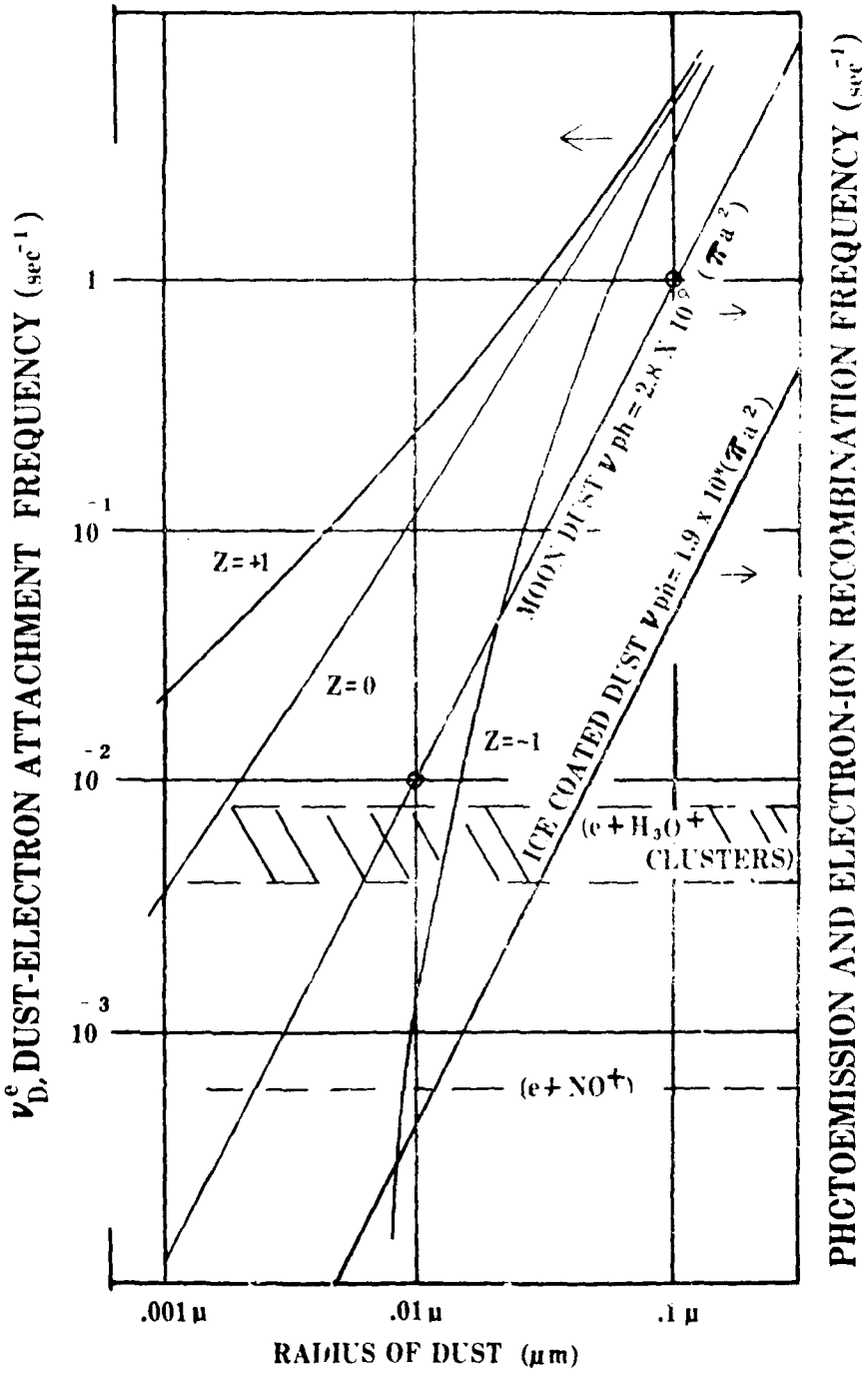

Figure 3. A plot showing the magnitude of ν_D^e, the dust-electron attachment frequency (left ordinate) for various radii of dust of charge Z = -1, 0, +1. The electron-cluster ion and electron-nitric oxide ion recombination frequency is plotted on the right ordinate. An electron density of 10^3 cm^{-3} is assumed. Values of photoemission frequencies for various radii of moon dust and ice-coated moon dust are represented on the right ordinate.

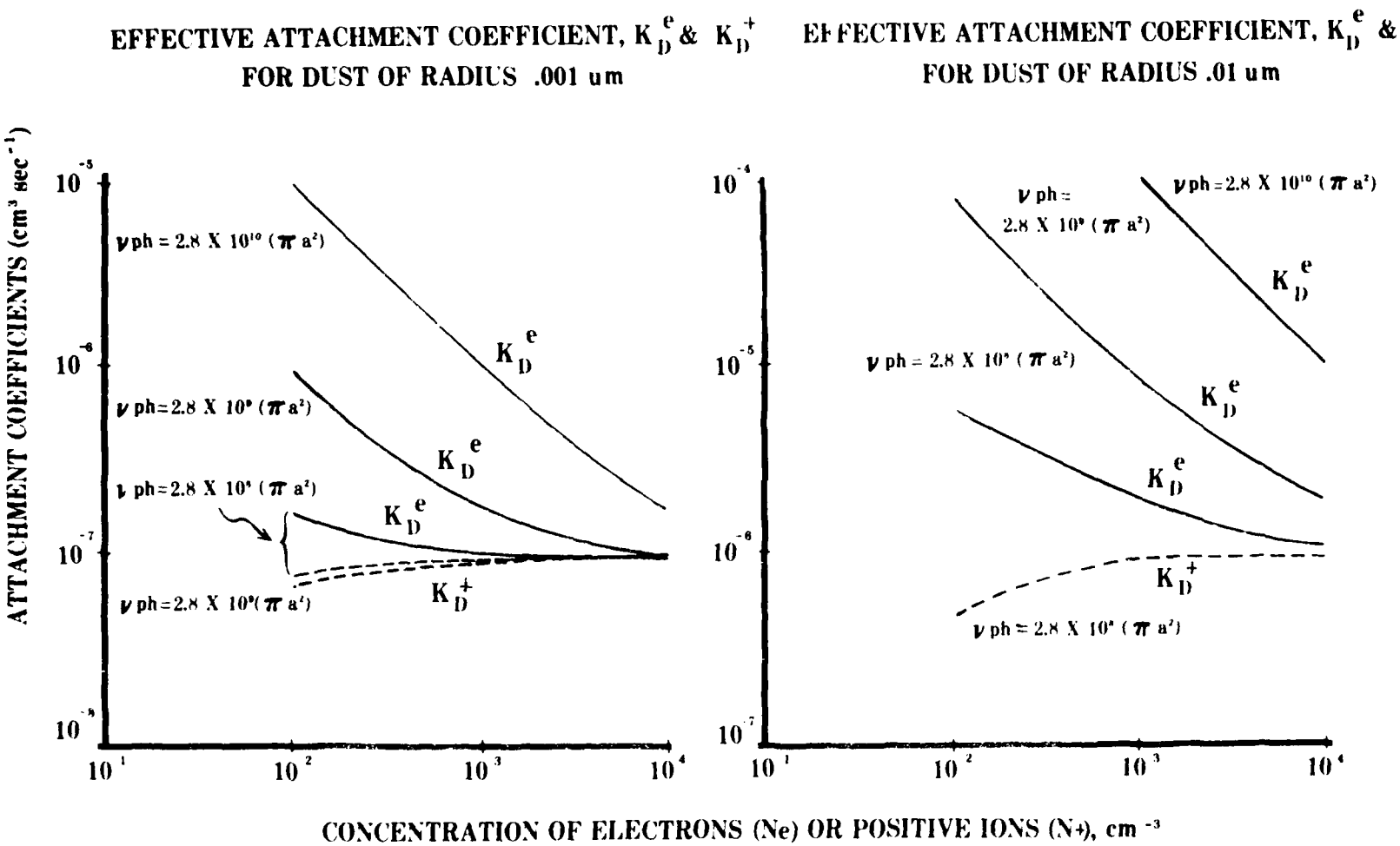

Figure 4. Values of the effective attachment coefficients of 0.001μm and 0.01μm dust to electrons, K_D^e, and positive ions, K_D^+, for populations of electrons and ions ranging from 10 to 10^4 cm^{-3}.

would depend on the rate of photoemission of the electrons from the negatively charged dust.

b. During twilight or nighttime, smaller particulates (radius <0.01μm) become negatively charged. Hence, in large numbers, small dust particles could compete effectively with ion-electron recombination and provide a means of maintaining (rather than neutralizing) negative charge. Referring to the right side of equation (6), the ν_D^e term will dominate the tendency to load the particles negatively; the photoemission term, ν_{ph}, will tend to counter this and may, if sufficiently large, keep the smaller particles neutral or even make them positive. At night, or for regions of low light levels, the ν_D^e term will dominate and drive a large fraction of the particulates negative.

c. Dust which is ice coated photoemits an order of magnitude less rapidly than uncoated dust. This difference would mean that during polar summer conditions with noctilucent clouds, a larger number of electrons could be attached to the ice coated dust than at other times of the year, resulting in a sharper reduction in the free electron density.

4.4 Effective Dust Attachment Coefficients

Figure 4 shows one instructive example of the derived ν_D^e and K_D^+ values for 0.001μm and 0.01μm size dust plotted against electron density (N_e) and three photodetachment values. The abscissa for figure 4 is N_e (or N_+ since, by the requirement of charge neutrality, they must be held equal in the calculations). Immediately apparent from this graph is the great variability of K_D^e and K_D^+ with changing electron and positive ion densities and photodetachment. Specifically: (a) K_D^e exceed K_D^+ since electrons move 300 times as fast as positive ions and hence have a higher collisional frequency. (b) Higher values of photodetachment rates, ν_{ph}, result in higher K_D^e values since photodetachment tends to keep a particle uncharged (or positively charged), which means that impacting electrons will not feel a repulsive force. (c) As the numbers of ambient electrons and ions increase, the K_D^e and K_D^+ values converge to a common asymptote because the extreme ambient densities negate the effects of photodetachment. However, the fact that K_D^e and K_D^+ are so highly dependent upon ambient electron and ion densities requires that they be entered iteratively into computer codes, a less efficient procedure than using a set of k_D^e and k_D^+ for the individual charging reactions for dust.

5. THE EFFECTS OF DUST ON D REGION CHEMISTRY AND CHARGE BALANCE THROUGH THE USE OF A FULL CHEMICAL CODE

5.1 Runs at 80 km with Variable Dust Concentration and Photodetachment Frequencies

The results of the previous sections have been organized as piecewise calculations designed to gain insight into the behavior of dust in the mesosphere. In this final section, dust is entered as a full partner into an electron-ion reaction set to quantify the effects on the charge balance. The treatment of this topic is not exhaustive, but it is included in this report to establish the qualitative nature of the effect of dust in the D region. The chemistry reaction set is that of DAIRCHEM,[30] and the dust attachment coefficients used are $k_n^e = 1.2\ (-4)$ cm^3 s^{-1} for 0.01μm dust (table 1). Dust is entered in several concentrations: 0, 100, 1000, and 10,000 cm^{-3}, and the resulting values in electron density, NO^+, and the sum of the hydrated water cluster ions are listed in table 3, examples 1 through 4. In these examples the photodetachment value is held at the value derived from Willis et al,[25] 2.8 x 10^9 (πa^2) s^{-1}, and the dust concentration is varied by 4 orders of magnitude. A second set of examples, 5 through 7, tests the results using photodetachment rates which vary from 0.01 to 100 times the above value.

Examples 1 through 4 show that as the dust population increases from 0 to 1000 cm^{-3} and 10,000 cm^{-3}, the electron density decreases from a high of 453 to 191 and 54 cm^{-3}, respectively. Sixty-four percent of dust remains uncharged at high concentrations (10,000 cm^{-3}), and 28 percent is negatively charged. Besides the effect of reducing the electron density, another major change is the marked increase in water cluster ion population. This increase is due to the removal of electrons which would recombine with the cluster ions directly (that is, due to the lower ion-dust recombination rate). Total charge neutrality is maintained in the chemistry of these runs, although only a small subset of positive ion species entered in DAIRCHEM is listed in this table. (For example, nitric oxide clusters and other minor positive ion species are omitted.)

Examples 5 through 7 show that the variation in photodetachment is significant in modifying the free electron density. High photodetachment rates drive the dust more positive and hence release electrons. For an increase in photodetachment of 4 orders of magnitude, the free electrons increase from 118 to 749 cm^{-3}. For the most probable value of photodetachment (Willis' value of 2.8 x 10^9 πa^2 cm^2 s^{-1})[25], the electron density increases more than 60 percent over its value with no photodetachment.

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[30] D. W. Hoock and M. G. Heaps, 1978, DAIRCHEM: A Computer Code to Model Ionization-Deionization Processes and Chemistry in the Middle Atmosphere. Users Manual, US Army Atmospheric Sciences Laboratory, White Sands Missile Range, NM.

[25] R. F. Willis, M. Anderegg, B. Feuerbacher, and B. Fitton, "Photoemission and Secondary Electron Emission from Lunar Surface Material," in Photon and Particle Interactions with Surfaces in Space, Proceedings of the 6th Eslab Symposium, Noordwijk, The Netherlands, 26-29 September 1972. R. J. L. Grand, Ed., D. Reidel Publishing Co., Dordrecht-Holland/Boston - USA.

TABLE 3. VARIATION IN ELECTRON DENSITY AT 80 km DUE TO VARIABLE DUST CONCENTRATION AND PHOTODETACHMENT. RADIUS = 0.01 μm. DAIRCHEM COMPUTER MODEL MIDLATITUDE CASE. ν_{ph} IS LISTED FOR ZERO OPTICAL DEPTH AND AN UNCHARGED PARTICLE.

Photodetachment, ν_{ph}	Example	Dust Entered	Uncharged Dust	Dust(+)	Dust(-)	Electron Density	NO^+ Ion Density	Sum Water Cluster Ions
	1	0	0	0	0	453	102	207
2.8×10^9 (πa^2)	2	100	23	1	76	406	103	232
	3	1000	384	13	603	191	111	507
	4	10000	6423	759	2818	54	117	1818
2.8×10^7(πa^2)	5	1000	11	10^{-3}	989	118	114	823
2.8×10^9(πa^2)	6	1000	384	13	603	191	111	507
2.8×10^{11}(πa^2)	7	1000	524	443	33	749	94	118

In each set of test runs the NO^+ population is comparatively stable. In addition to the reactions listed below, a set of attachment reactions of positive ions to neutral, negatively charged, and positively charged dust was also used.

$$D + e \rightarrow D^- \quad (8)$$

$$D^+ + e \rightarrow D \quad (9)$$

$$D^- + h\nu \rightarrow D + e \quad (10)$$

$$D + h\nu \rightarrow D^+ + e \quad (11)$$

5.2 Comparison of the Effect of Four Dust Profiles ($r = 0.01\mu m$ and $r = 0.001\mu m$) upon D-region Ion Chemistry.

As a more graphic test of the possibility of dust modifying the electron profiles of the D region, four dust distributions were included in the DAIRCHEM code, and computations were made at a series of altitudes. As a basis of comparison for the modeling, a standard type of observed electron density profile from daytime, $\chi = 60°$ at Wallops Island, VA, in December is presented. Secondly, the electron density profile computed by DAIRCHEM with no dust included is plotted. The other curves are computed electron densities using $0.01\mu m$ dust at constant concentrations of 100 and 1000 cm^{-3} and $0.001\mu m$ dust at concentrations of 1000 and 5000 cm^{-3}. These values bracket the estimates generally accepted for dust concentrations in that region. The values are tabulted in table 4. A photodetachment value of $2.8 \times 10^9(\pi a^2)\ s^{-1}$ was used for dust for all runs.

TABLE 4. SELECTED DUST PROFILES

Altitude (km)	A	B	C	D	E
	No Dust	0.01μm		0.001μm	
100	0	100	1000	1000	
90	0	100	1000	1000	5000
85	0	100	1000	1000	
80	0	100	1000	1000	5000
75	0	100	1000	1000	
70	0	100	1000	1000	5000
60	0	100	1000	1000	

The dust profiles used as input to the code represent a plausible range of values. However, since the distribution of dust in the upper mesosphere is a very open question, these profiles were felt to be adequate in a preliminary demonstration of the DAIRCHEM results to dust, particularly the charge

balance. A "step" profile of dust (i.e., a sharp ledge of dust at a given altitude) has been conjectured in the literature. This effect may easily be seen by the comparison, at any altitude, of the dust case with the no dust case.

The results of these inputs to DAIRCHEM are plotted in figure 5. Curve 1 is the Wallops Island observation of electron density for December with χ = 60°. This profile shows the abrupt change in slope at 82 km that is commonly observed in many electron density profiles in the D region under quiet conditions. Curve 2 is the computed electron density profile using DAIRCHEM with no dust. Note the divergence of these two curves at 87 to 75 km. Curves 3 and 4 are 0.01μm dust at concentrations of 100 cm^{-3} and 1000 cm^{-3}. A reduction in electron concentration is apparent at 85 to 78 km, particularly for the 1000 cm^{-3} distribution. Below 78 km, the reduction in electron density exceeds the observed curve 1, possibly suggesting that dust may be confined in a relatively narrow ledge. For example, the best fit using 0.01μm dust would be in excess of 1000 cm^{-3} from 85 to 77 km and dropping to about 200 cm^{-3} by 70 km. Curves 5 and 6, 0.001μm dust at 1000 and 5000 cm^{-3}, bracket the observed values of electron density down to 77 km. Optimum concentration for 0.001μm dust would be 3000 to 4000 cm^{-3}. Below 77 km the reducing action of this dust is clearly excessive, thus suggesting values near 200 cm^{-3} at 70 km.

Although these calculations were run at each 5 km in altitude and with a limited number of cases, the results clearly show that a judicious choice of dust concentration can allow the modeled electron density to reproduce the heretofore "anomalous bite-outs" in electron density often observed around 80 km. The controlling rule is: if the particle concentration equals or exceeds the model electron density computed by vapor phase reactions alone, such as in the case below 85 km, a reduction to observed values is apparent. Most runs of DAIRCHEM indicate the best fit is with a variable concentration of dust with altitude.

6. SUMMARY AND CONCLUSIONS

The purpose of this report has been to examine, through a series of piecewise calculations and comparisons, the importance of electron-dust attachment to D region chemistry, particularly in competition with electron-ion recombination. In the course of the argument, we have examined the preferred charge loadings on a population of dust and generated two different (but related) sets of coefficients for electron or positive ion dust attachment calculations. The discrete attachment coefficients, k_D^e and k_D^+, should generally be used in chemical computer codes since they result in knowledge of the precise charge loading of individual dust particles.

Dust of radius less than 0.01μm acquires only one electron under mesospheric conditions. Dust of radius 0.1μm may acquire as many as five or six resident electrons, and in this size range photodetachment does not perturb the distribution of charge loading too greatly. The size of dust therefore seems to predetermine a unique charge loading for each dust particle over a wide range of photodetachment values. An accurate measurement of dust size and

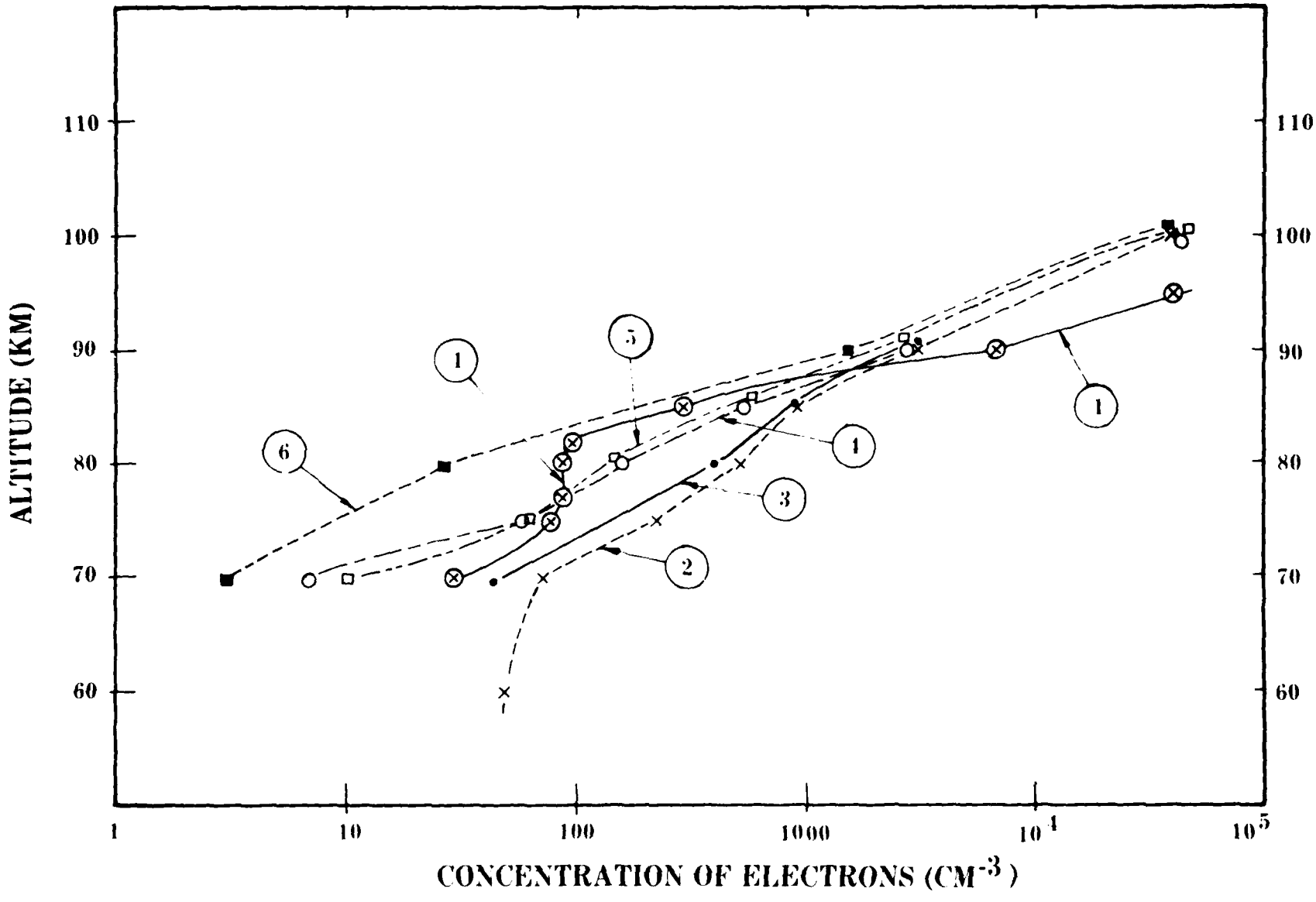

Figure 5. A plot of observed electron densities at Wallops Island, VA, for December (curve 1), model computation by DAIRCHEM from the case of no dust (curve 2), 0.01μm dust concentrations of 100 cm^{-3} and 1000 cm^{-3} at all altitudes (curves 3 and 4), and 0.001μm dust at concentrations of 1000 cm^{-3} and 5000 cm^{-3} (curves 5 and 6).

concentration could permit one to reasonably estimate its effect upon the chemistry due to this unique property. The full understanding of the role of dust, however, requires the use of the ion kinetic reaction scheme.

This report ameliorates some uncertainty in photodetachment rates by running a set of calculations of DAIRCHEM using ν_{ph} orders of magnitude lower and higher than the value used by previous researchers and showing resultant stability in the computed electron densities. Nevertheless, the previous value, obtained from moon dust, seems to be the most logical for use in further D region chemistry runs. Based upon the limited computations made for this study, dust indeed does appear to compete strongly for electrons, particularly at dust concentrations of around 1000 cm^{-3}.

Figure 5 represents the most advanced set of calculations of this report and demonstrates that it is possible, with a proper choice of a dust profile and size, to approximate several heretofore "anomalous features" in electron density profiles.

The concentrations of dust included here for use at 80 km were consistent with reports from the literature. The next step in pursuing this topic must be the analytic derivation of a dust size concentration profile starting, most logically, with meteor flux estimates at 150 km. This present model has been run strictly for daytime chemistry and has purposely neglected nighttime processes and the attachment of negative ions to dust at night. Until this other important step is taken and an extensive series of runs is made, a description of the role of dust in D region chemistry will remain incomplete.

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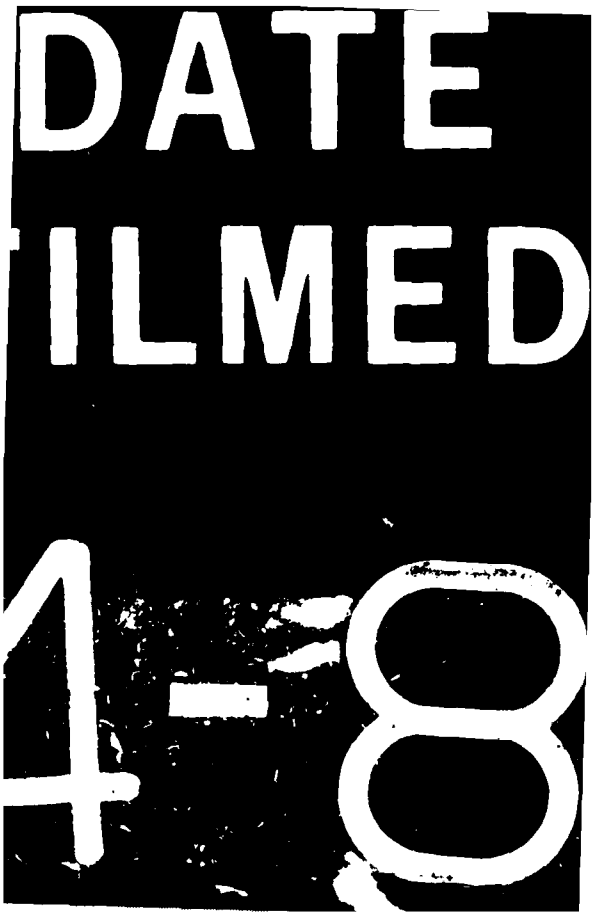
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